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Fuel-Neutral Studies of Particulate Matter Transport Emissions

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Pacific Northwest National Laboratory

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Overview

Timeline

- Start - FY09
- Propose to continue while making progress

Budget

- Funding received in FY14 - $200K
- Planned request for FY15 - $200K

Barriers

- Barriers addressed for enabling of high-efficiency engine technology:
  - B. Lack of cost-effective emission control
  - C. Lack of modeling capability for combustion and emission control
  - F. Lack of actual emissions data on pre-commercial and future combustion engines

Partners

- General Motors Company - provide project guidance, provide hardware to ERC
- Engine Research Center at University of Wisconsin, Madison - host and operate test engines

Relevance and objectives

Overall objective: Enable application of future high-efficiency engine technology

Barrier: Lack of actual emissions data on pre-commercial and future combustion engines
Objective: Comprehensive particulate characterization with single-cylinder test engines, guided by industry

Barrier: Lack of cost-effective emission control
Objective: Seek to shorten development time of filtration technologies for future engines by improving fundamental understanding of how filter media properties impact back-pressure and filtration efficiency

Barrier: Lack of modeling capability for combustion and emission control
Objective: Develop modeling approaches relevant to the likely key challenge for SIDI filtration – high number efficiency at high exhaust temperatures (implying little soot accumulation in filters)

Approach - Background

- When this project began, very little was known about soot production in SIDI and other advanced gasoline engines
  - How much PM will be produced with various fuels and operating conditions?
  - How will particulate properties (size distribution, morphology, composition, etc.) differ from Diesel soot?
- Filtration of particulates from advanced gasoline engines may entail unique challenges
  - Due to high exhaust temperatures and relatively low soot loadings, SIDI filter systems may not accumulate soot cakes, which may limit particulate removal efficiency
  - Fuel efficiency is likely to be very sensitive to exhaust system back-pressure
- System design engineers need better modeling tools to predict filter back-pressure and removal efficiency across the particle size spectrum and understand how filter media properties affect performance
  - Unit collector models have been widely used for design of DPFs
    - Classical approach is based on analytical forms for flow around isolated spheres, adjusted to fit experimental data from beds of granular particles
    - Properties of packed beds of mono-sized spheres are easily represented by a single length scale in this scheme
    - Commonly used exhaust filter media, like cordierite and aluminum titanate bear very little resemblance to loose granular beds
    - Models could nevertheless be tuned through a representative collector diameter to match total mass removal for design of DPFs, but accurate number efficiency prediction as a function of PM size is more challenging

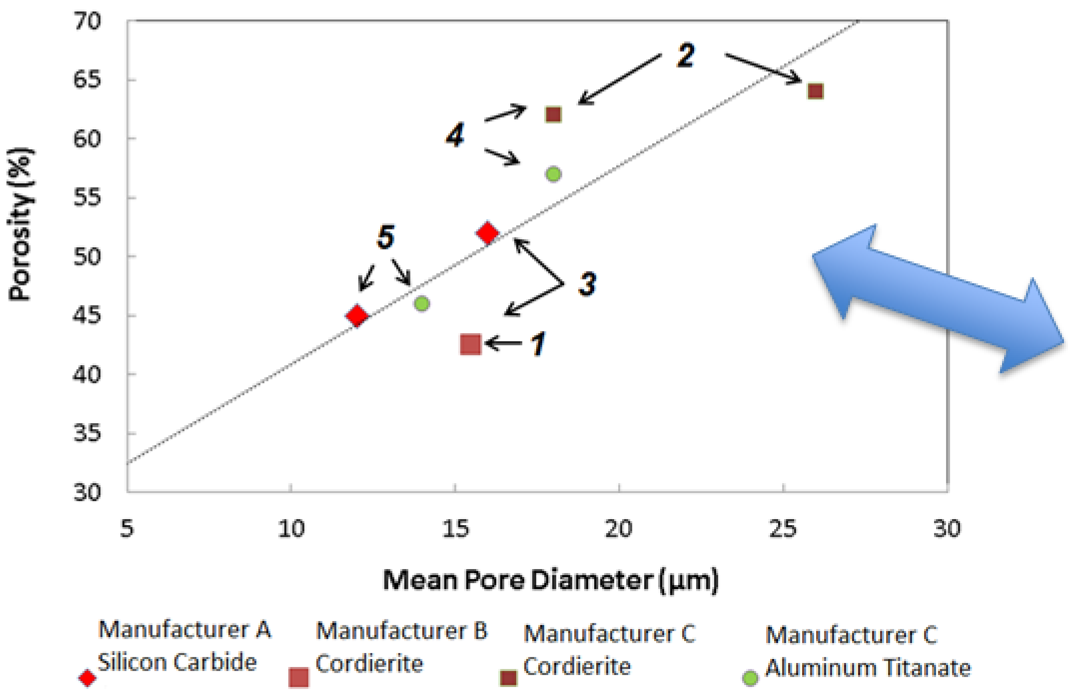
Approach

- Particulate characterization
  - Two extensive cooperative experimental campaigns were carried out with the University of Wisconsin ERC, including dozens of experiments at various relevant engine operating points in several test engines with multiple fuel blends
  - Standard and advanced characterization techniques including SMPS, SPLAT II, APM, TEM, HRTEM, FTIR-ATR, and XPS *were used together for a more complete picture of particulate populations*
  - Analysis has resulted in a very large dataset encompassing many characteristics of advanced gasoline particulate populations: size distributions, aggregate shape, primary particle size, composition, density
  - Data has provided new insights into the relationship between PM properties and engine operating conditions and fuels, guiding development of engine control strategies, planning of future experiments, and development of aftertreatment solutions
- Filter analysis and modeling
  - Techniques are being developed to characterize the actual 3D structure of exhaust filtration media from micro X-Ray CT data and understand how structure impacts performance
  - More sophisticated modeling approaches are being explored to improve a-priori predictions of filter performance in a given application
    - How many parameters are necessary to completely predict actual filter performance?
    - Can they be tied to measured dimensions or properties?
  - Filter efficiency experiments, including the Exhaust Filtration Analysis (EFA) platform at the University of Wisconsin, Madison ERC are being used to collect data with a wide variety of filter substrates

Micro X-Ray CT analysis of a variety of samples

Initial Samples	Manufacturer	Mercury Porosity (%)	Mercury Median Pore Size (microns)	Length Scanned (mm)
Substrate 1	B	62.8	22.6	19.4
Substrate 2	B	54.4	18.8	10.3
Substrate 3	D	52.4	17.2	10.5
Substrate 4	D	62.2	20.6	10.5
Substrate 5	D	55.5	16.6	11.2

- High res CT and mercury porosimetry data obtained and analyzed for 5 initial cordierite samples covering a range of porosities and pore sizes



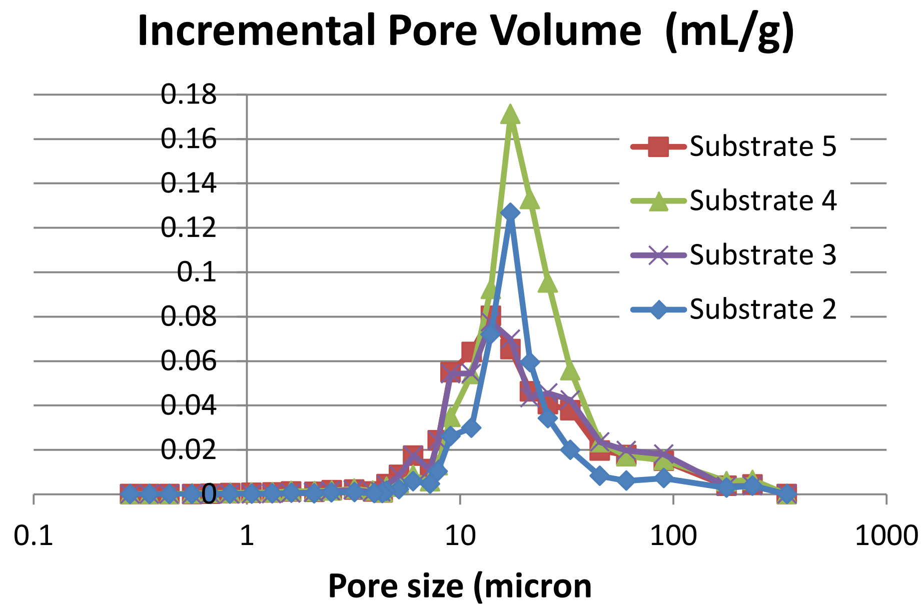
- CT data is being obtained for 7 additional samples to support modeling of EFA experimental matrix:

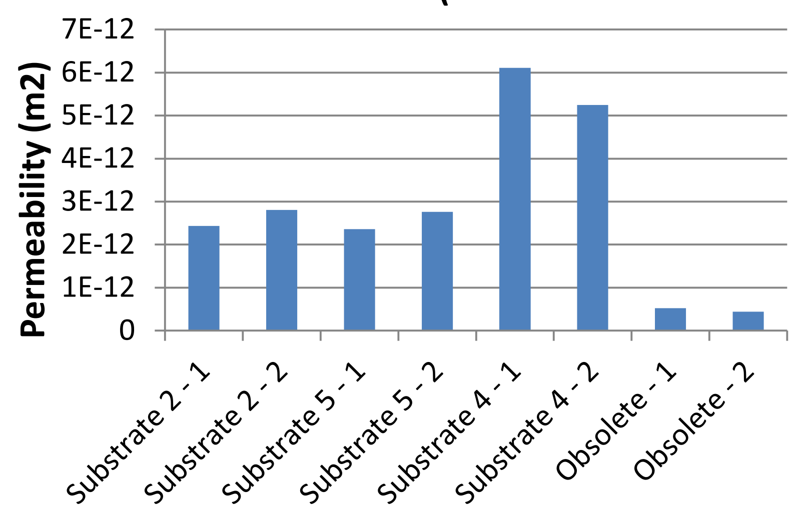
1. Scoping experiments
2. Effect of pore size
3. Effect of porosity and material
4. Effect of porosity and material
5. Effect of pore size and material

Alloy Foam filter samples have also been obtained from a fifth manufacturer for initial evaluation

Mercury porosimetry data

- Hg porosimetry data is easily obtainable
- Pore sizes may not correlate exactly to any physical dimension
- Narrow pore size distributions are thought to give the highest filtration efficiency at a given pressure drop
- CT data gives three-dimensional picture of pore structure with a resolution of 1.6 micron





Technical Accomplishments

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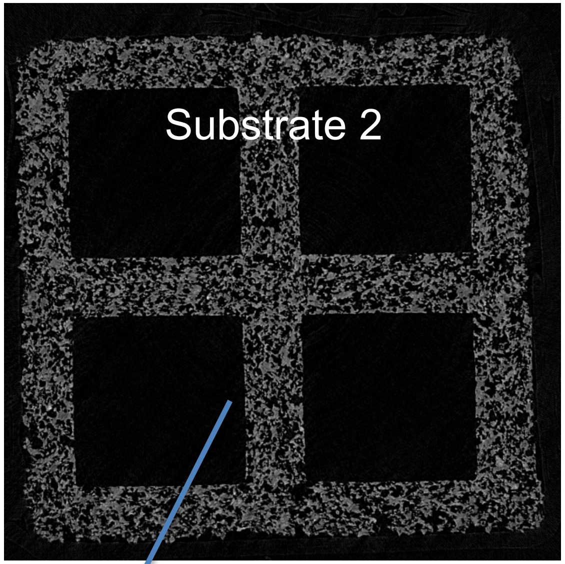
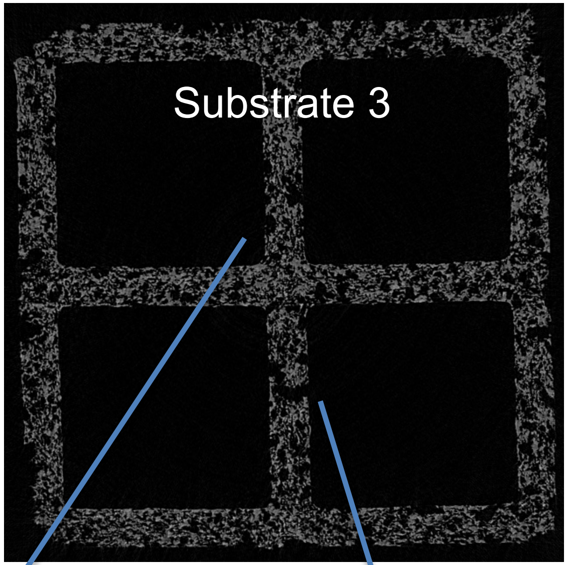
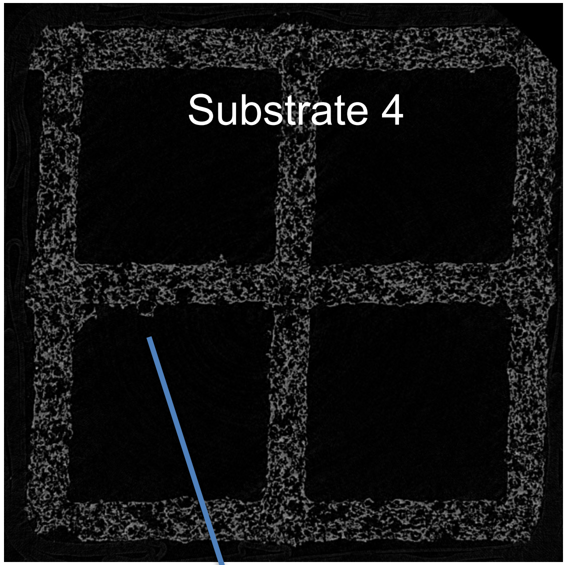
Examples of raw CT images







Narrower pore size distribution apparent in this substrate

Some large pores traverse much of the wall thickness

Occasional large-diameter paths through the wall

Some substrates had much rougher walls, some had 'blisters' near the surface

- Cordierite substrates from different manufactures have visibly different structures, even when porosity and mean pore size are comparable

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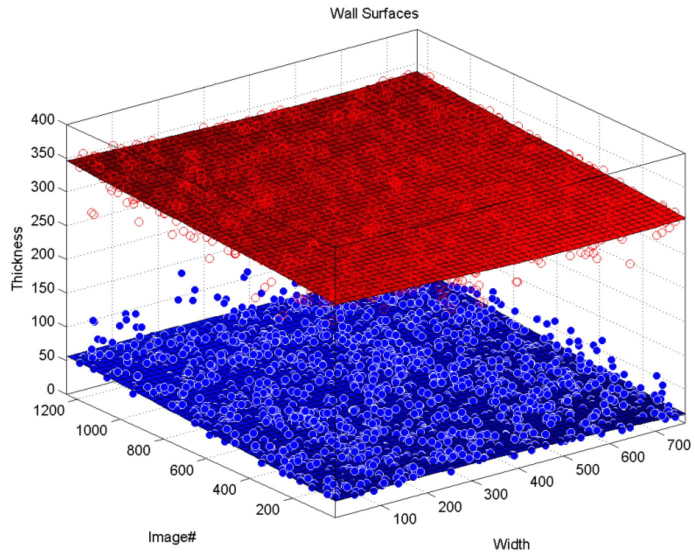
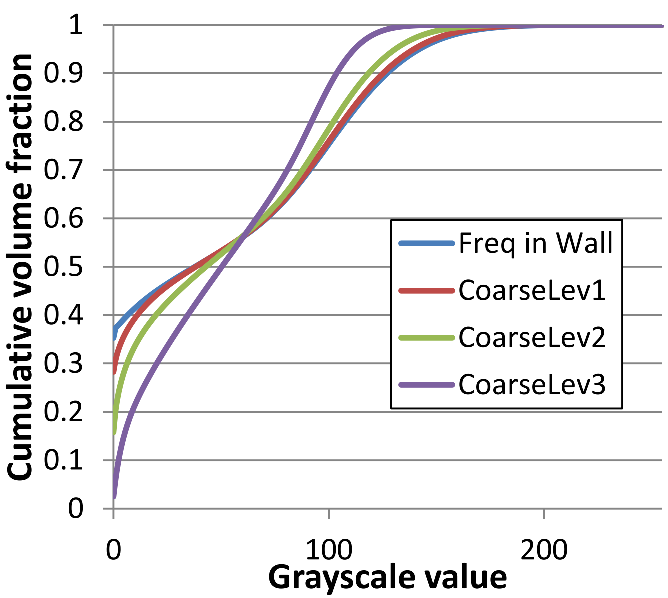
Automated analysis of CT data

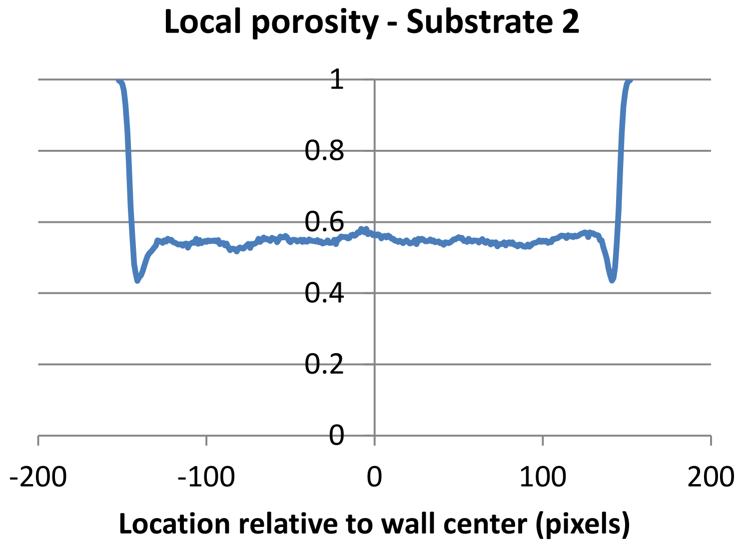
- Average wall surfaces were fit by third order polynomials
- Internal locations could then be interpolated between the two surfaces to scan through the wall thickness
- Grayscale histograms were obtained for original resolution and for three levels of coarsened geometries to allow thresholding to match measured porosities at various resolutions
- Higher or lower thresholds could be chosen to create similar structures with various porosities





Porosity scans across wall thickness

- All of the cordierite samples scanned exhibited low porosity regions near the wall surfaces
- Lower porosity at wall surfaces may not have a major effect on pressure drop or clean filtration efficiency, but probably affect transition to cake filtration and pressure drop response with even small amounts of soot accumulation
- For matching total porosity and calculating wall thickness, wall 'surfaces' were considered to be the locations of minimum porosity
- Wall thicknesses varied from nominal values



	Nominal Thickness (mil)	Nominal Thickness (microns)	Observed Thickness (microns)	Difference
Substrate 1			365	
Substrate 2	17	430	451	+ 5%
Substrate 3	12	300	277	- 8%
Substrate 4	12	300	300	0 %
Substrate 5	12	300	281	- 6%

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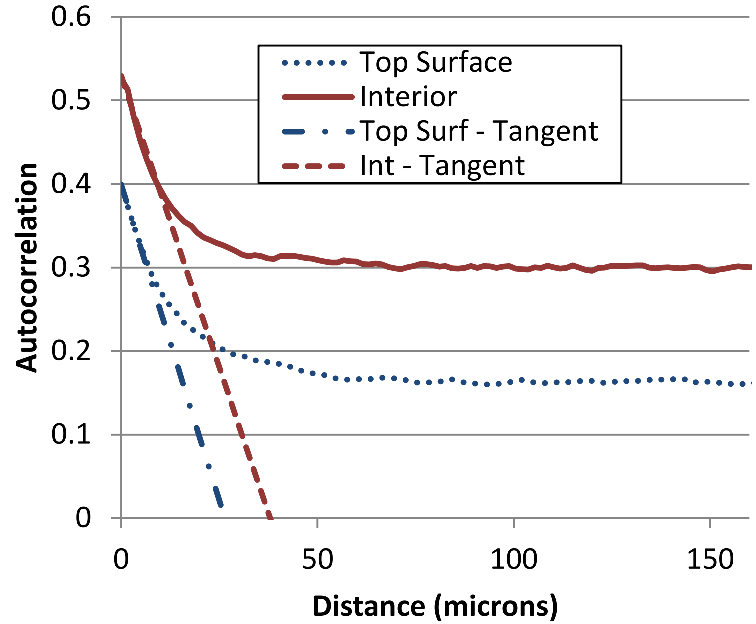
CT analysis - length scales

- Autocorrelation distributions are easy to calculate and are a standard measure of characteristic length scales in random heterogeneous media
- Intercepts of short-distance tangent lines have been used as one metric to find equivalent collector diameters in SiC filters [1]
- Like porosity, intercept distances are significantly different between the wall surface and interior

Void Autocorrelation Distributions - Substrate 3

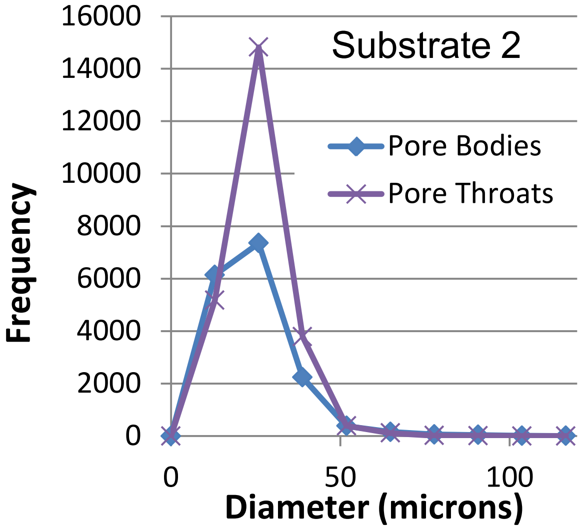


	Void Intercept (microns)		Solid Intercept (microns)	
	Surface	Interior	Surface	Interior
Substrate 1	31.04	40.16	28.32	21.76
Substrate 2	27.84	39.04	33.44	27.2
Substrate 3	26.08	37.44	37.28	18.24
Substrate 4	32.96	47.36	28	15.84
Substrate 5	29.6	44	32.8	18.4

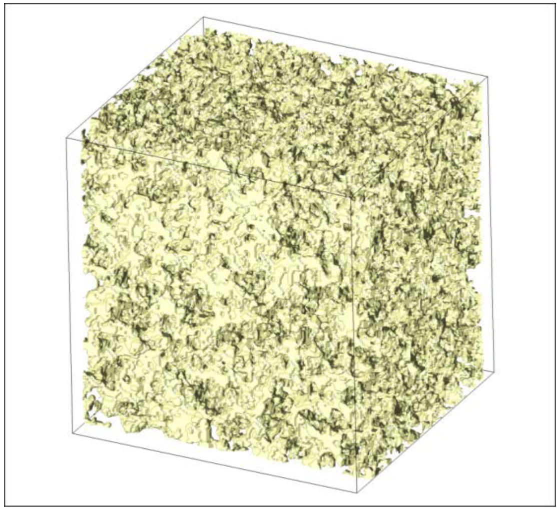
[1] Fukushima, S., K. Ohno, N. Vlachos, and A.G. Konstandopoulos, "New Approach for Pore Structure and Filtration Efficiency Characterization". *SAE*, 2007. 2007-01-1918.

Analysis by maximal inscribed spheres

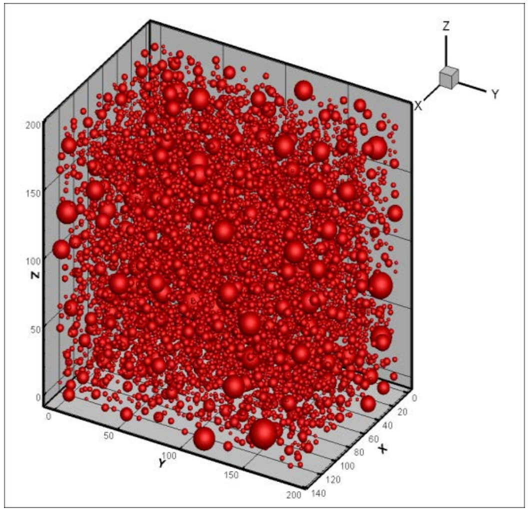
- The Maximal Inscribed Spheres (MIS) method has been used to analyze complex 3D pore volumes in CT data from rock samples, including simulation of mercury porosimetry data
- This technique can differentiate between pore bodies and pore throats and quantify properties like pore connectivity
- A prototype exhaust filter MIS program has been developed and applied to segments of three of the substrate sample reconstructions at 3.2 micron resolution
- The approach seems promising, but further evaluation and comparison to other data are still needed



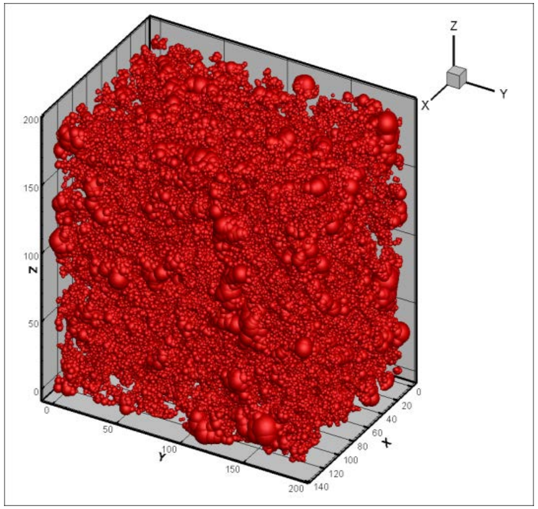
3D reconstruction - Substrate 2



Master spheres



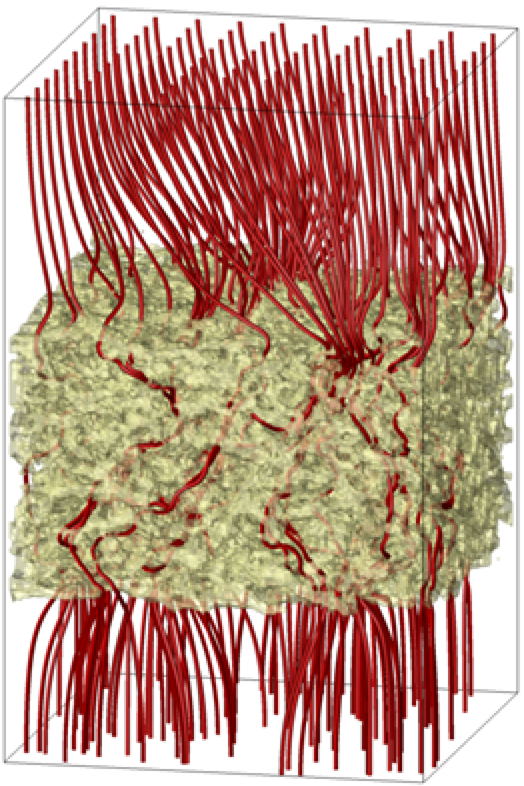
Sphere clusters



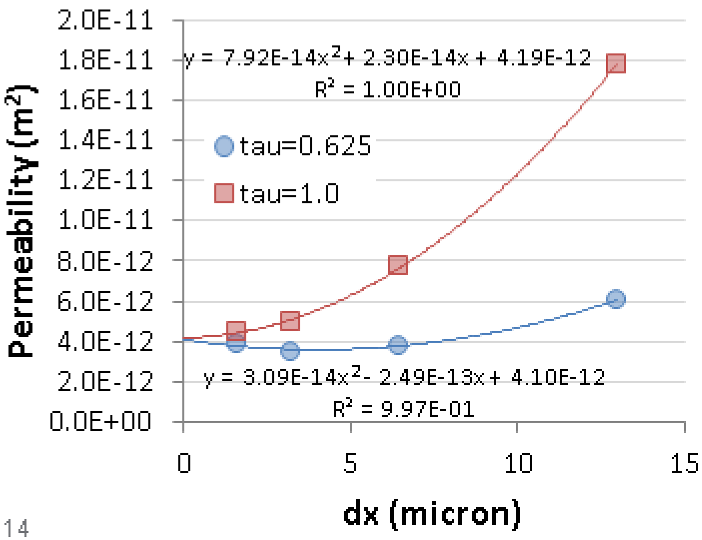
Lattice-Boltzmann flow simulations

- 3D reconstructions also allow pore-scale flow and transport simulations
- Discretization studies were carried out to determine required resolutions
- Dozens of LB simulations were run for each substrate to characterize Representative Equivalent Volume (REV)
- Calculated permeabilities agreed well with experiments

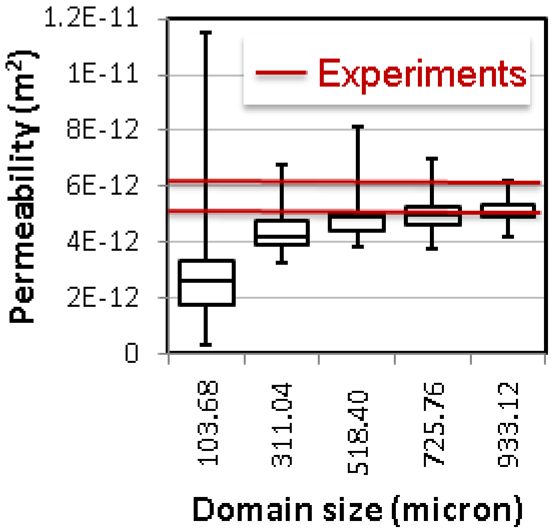
LB flow field - Substrate 4
dx = 3.24 micron
518 x 518 micron wall area



Discritization study - Substrate 4

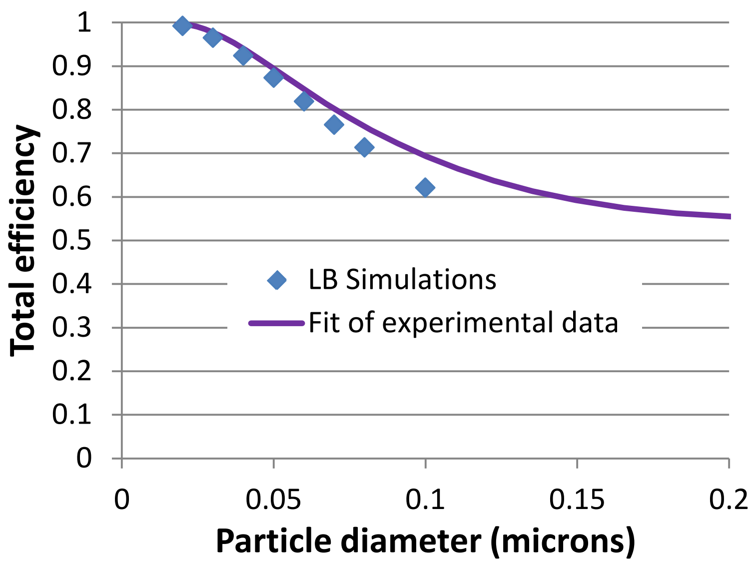


REV study - Substrate 4

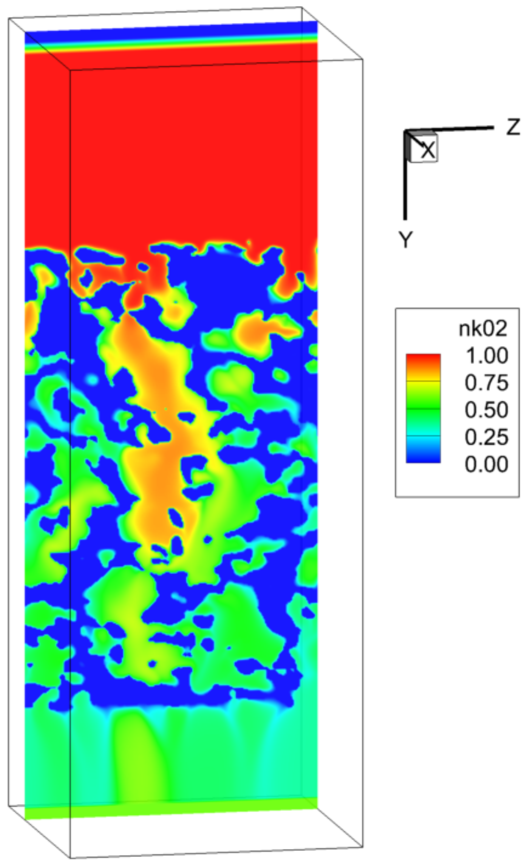


Lattice-Boltzmann filtration modeling

- Preliminary filtration simulations have been conducted with several of the substrate reconstructions using an Eulerian particle field representation
- Focus is on the diffusion efficiency term dominant for small particles
- This approach has been used to develop new forms of the diffusion efficiency relationship - tuned to specific porous media over a range of size scales and porosities
- Suitability of this method for exhaust filtration modeling is still being evaluated



LB filter model - Substrate 2
dx = 1.62 micron
311 x 311 micron wall area
100 nm particle at room temperature



Technical Accomplishments

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Heterogeneous Multi-scale Filtration (HMF) model

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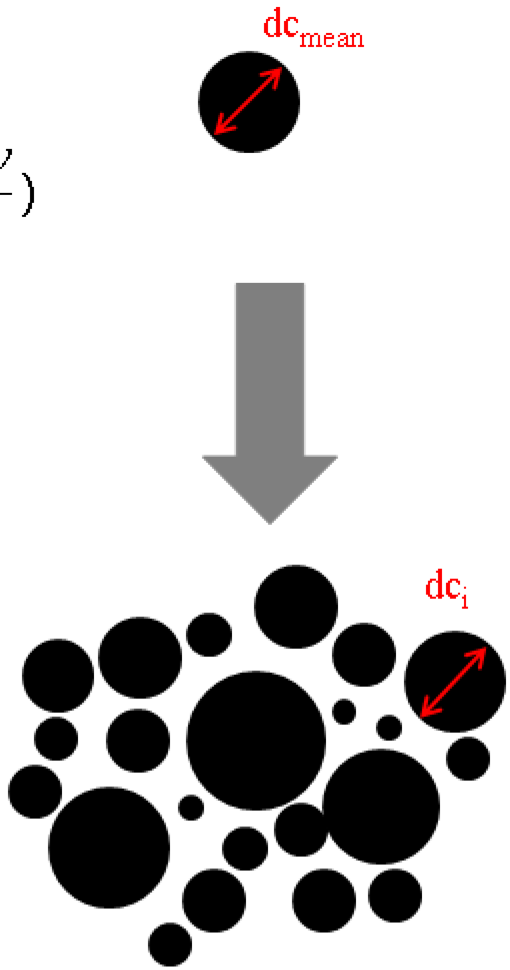
- Mean collector size (standard approach)
  - Mean pore size and mean porosity

$$\eta_{mean}(dp_i)=1-\exp\left(-\frac{3*\eta_{comb}(dp_i,dc_{mean})*(1-\epsilon_{mean})*w}{2*\epsilon_{mean}*dc_{mean}}\right)$$

- HMF
  - Use a cluster of collectors with different diameters to represent the complex porous structure
  - Pore size PDF and porosity distribution

$$\eta_i(dp_i,dc_i)=1-\exp\left(-\frac{3*\eta_{comb}(dp_i,dc_i)*(1-\epsilon_j)*w}{2*\epsilon_j*dc_i}\right)$$

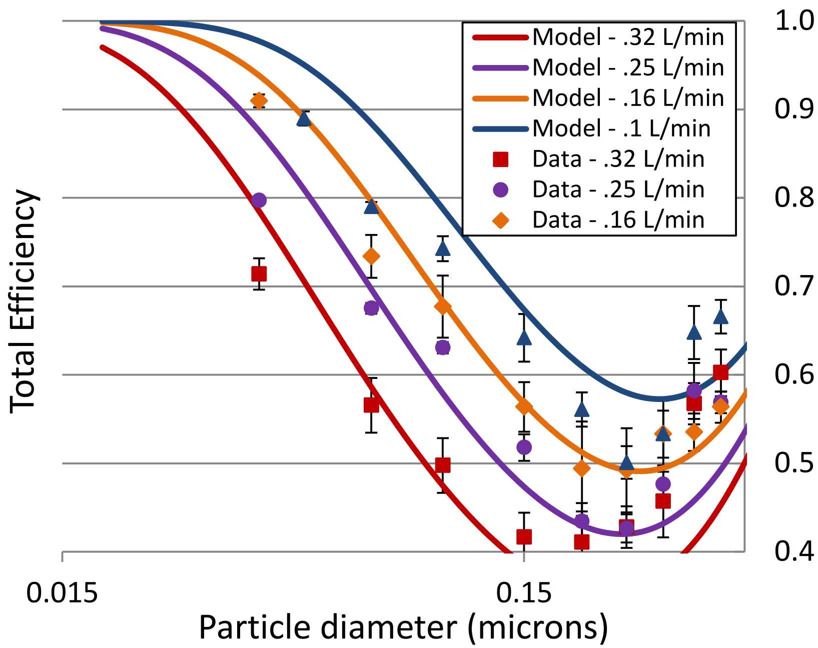
$$\boxed{\eta_{HMF}(dp_i)=\frac{\int \eta_i(dp_i,dc_i)\cdot dc_i^2\cdot pdf(dc_i)\,d(dc_i)}{\int pdf(dc_i)\cdot dc_i^2\,d(dc_i)}}$$



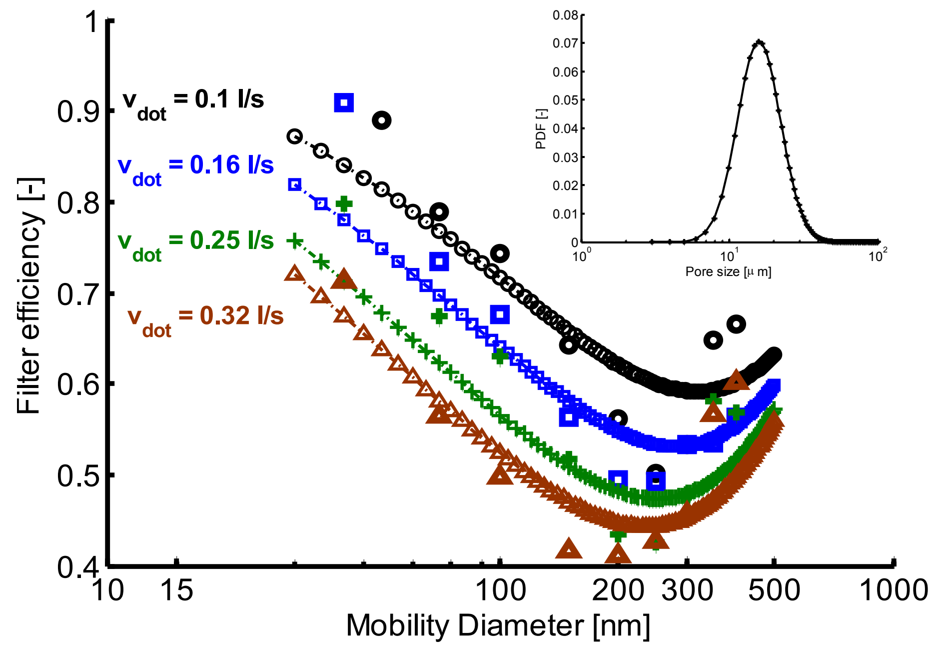
Heterogeneous Multi-scale Filtration (HMF) model

GM / UW-Madison Collaborative Research Laboratory

Mean collector with Long & Hilpert diffusion term



HMF model with log-normally distributed collectors



- Two improved models compared to PNNL single-channel filtration data
- Both mean collector with modified diffusion term and HMF models give better predictions of particle-dependent efficiency than the classical model

Technical Accomplishments

Exhaust Filtration Analysis (EFA) experiments

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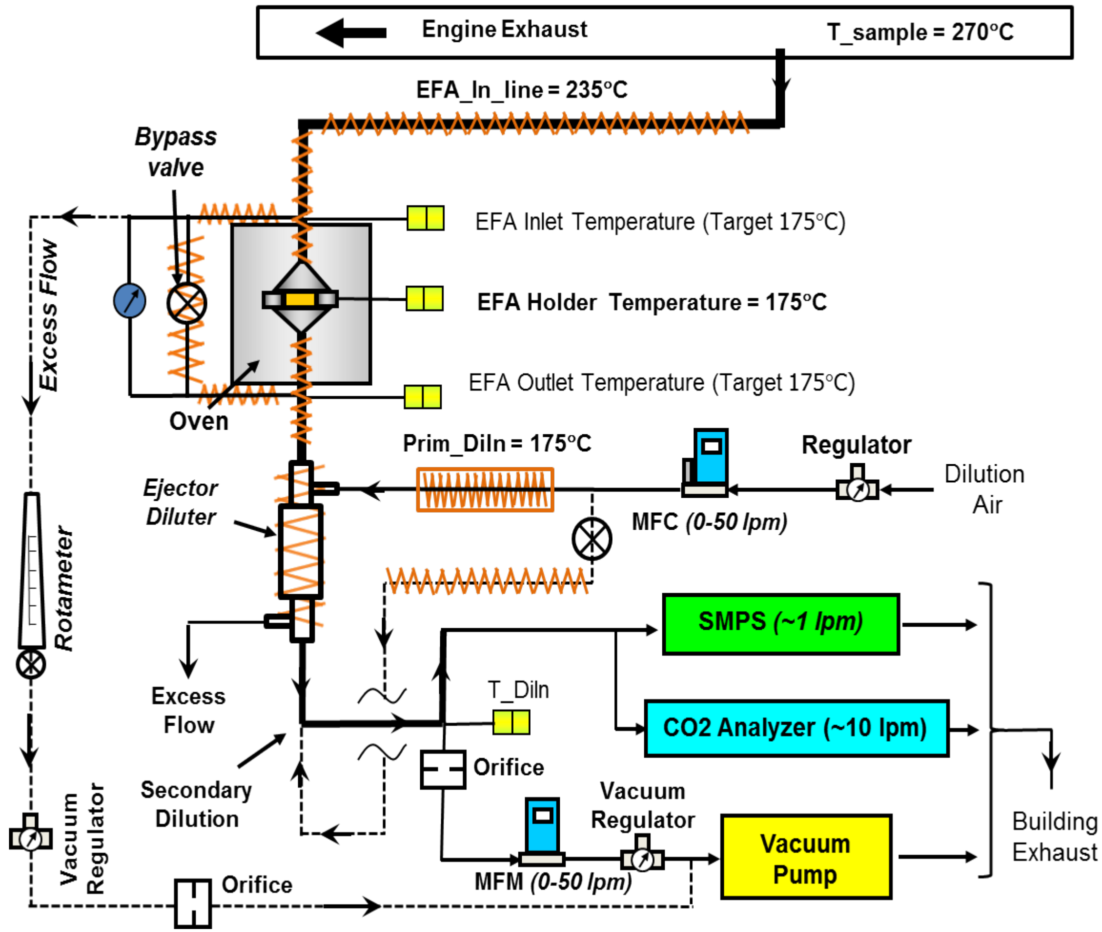
- Filtration experiments are being conducted with flat wafer samples and exhaust from single cylinder test engine
- Particulates are measured with Scanning Mobility Particle Sizer (SMPS) and Engine Exhaust Particle Sizer (EEPS)



See SAE-01-1158

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Exhaust characterization for EFA experiments

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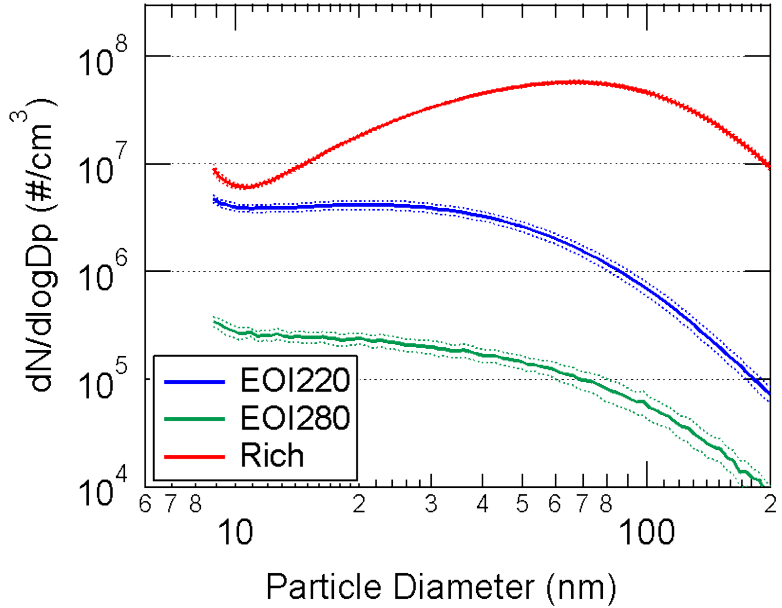
Engine operating parameters and resultant particulate emission numbers.

Condition	Speed	Injection Timing	IMEP (Gross)	AF Ratio	CA50	Total PM	Particles > 23 nm	Geometric Mean Diameter
	[rpm]	[˚bTDC]	[kPa]	[-]	[˚aTDC]	*1E05 [#/cm³]	[% of Total]	[nm]
EOI 220	2100	220	330 (±6)	15	8.0 (±0.5)	36.1	40	25
EOI 280	2100	280	350 (±5)	15	8.8 (±0.5)	2.21	40	25
Rich	2100	220	300 (±5)	13	6.7 (±0.5)	433	84	55

EOI 220 → Baseline Condition with injection during the intake stroke
EOI 280 → Early injection, near homogenous operation, low particle concentration, PSD shaped similar to baseline condition
Rich → Diesel-like size distribution



See SAE-01-1158

Technical Accomplishments

EFA filtration efficiency measurements
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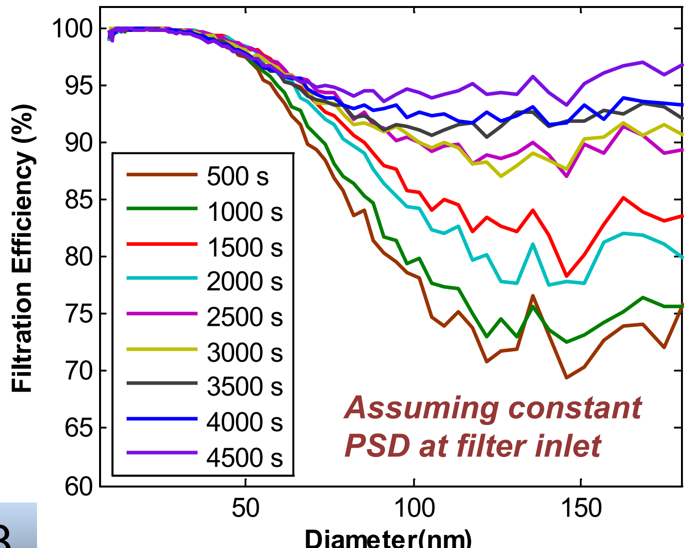
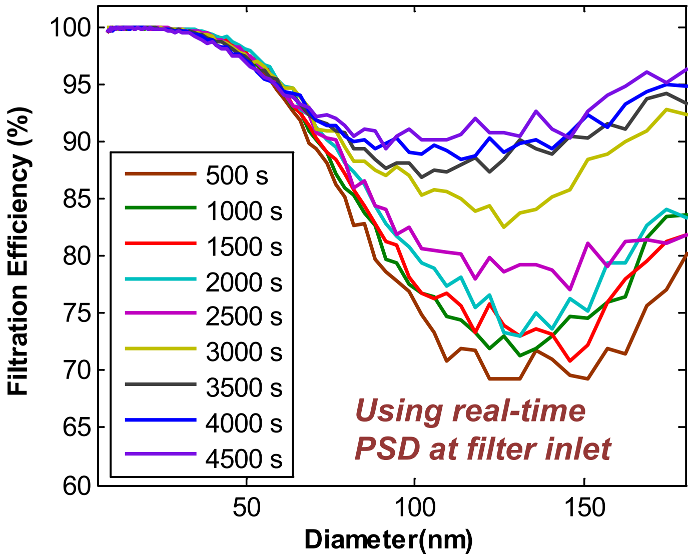
Condition	EOI 220
Filtration Velocity	2.6 cm/s
Temperature	175 ºC
Duration	5000 s

- Slow changes in engine-out particulates make it necessary to continuously adjust the measured inlet PSD using EEPS
- Trends show slow accumulation of particulates in filter which affects capture efficiency
- Efficiency changes more rapidly between 2000 and 3000 seconds of exposure
- Maximally penetrating particle size shifts from larger to smaller diameter over the course of the experiment

Filter Sample:
Cordierite
42.5% porous
15.5 μm mean pore diameter





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Application of HMF model to EFA experiments

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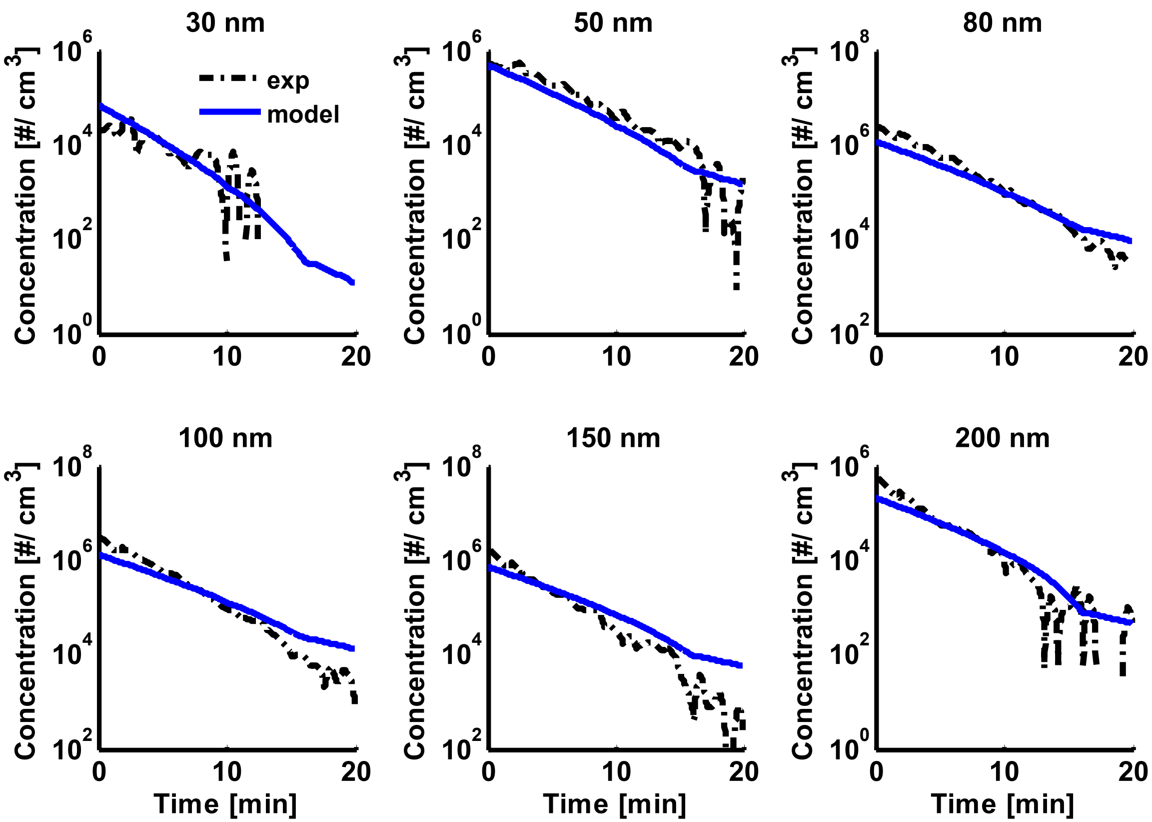
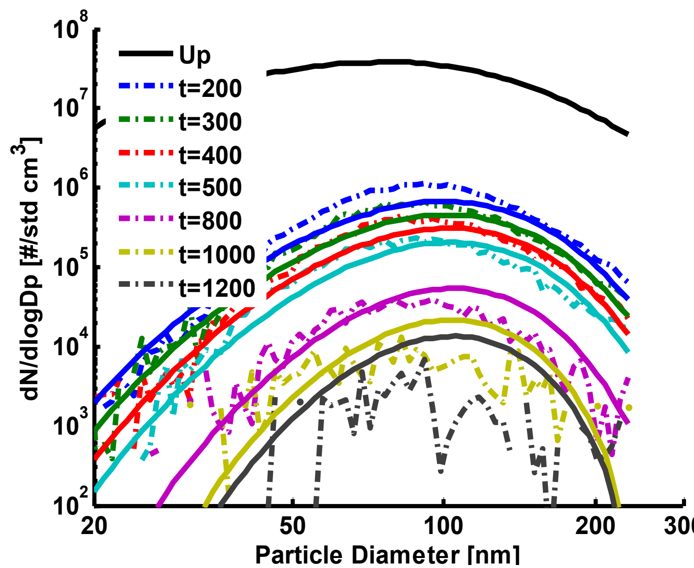




Gong, J. and Rutland, C., *"Filtration Characteristics of Fuel Neutral Particulates Using a Heterogeneous Multi-scale Filtration (HMF) Model"*, submitted to ASME 2014 Internal Combustion Engine Division Fall Technical Conference, Columbus, Indiana, USA

- Evolution of the particulate concentration penetrating the filter is well predicted by the HMF model across the whole range of particle sizes

FY13 Reviewer Comments

- "The reviewer criticized that the glaring weakness is generating or reproducing a membrane filtration technology which limits backpressure drop and has a high filtration efficiency for the small GDI particles."

  Experimental membrane filters have shown a great deal of promise, and received significant attention from us early in the project. However, these materials are not currently available, even for testing, and it is not clear that they will ever be commercially viable. The project team has chosen to focus on technologies that are likely to be available for application in the short to medium term, while developing fundamental understanding to assist in evaluation of future filter substrates.

- "The reviewer stated that there is no supplier involvement preventing the rating of outstanding."

  Instead of limiting explorations to a single supplier, the project team has elected to evaluate filter substrates from multiple suppliers. Four classes of materials from five different manufacturers spanning a very wide range of properties have been procured to date.

- "The reviewer indicated that the proposed future activities appeared reasonable. However, efforts should be made to include new filter materials such as membrane coated filters for PM studies and the effects of fuel sulfur and HC composition of E10 and E85 fuel blends."

  We have not seen evidence so far that fuel sulfur has a significant effect on particle number or morphology, but we will consider that in the future. The possible role of fuel HC composition in particulate formation has been discussed extensively by the project team, and considered in design of experiments.

Collaborations

- General Motors Company (Industry): Provide hardware, expertise, and operational guidance for engine experiments at the ERC. Advise on project direction and priorities.
- Engine Research Center at University of Wisconsin, Madison (Academic): Operate test engine - including shakedown tests, independent experiments, and cooperative experiments. Assist in analysis and publication of data. Develop improved device-scale modeling techniques.

Remaining challenges and barriers

- There is still a lack of comprehensive, high quality filtration data with a variety of filter substrates across the spectrum of particle sizes anticipated with advanced gasoline engines
- No exhaust filtration model has been demonstrated to reliably predict filtration efficiency as a function of particle size a-priori
- Engine technology and combustion strategies continue to evolve and fundamental questions about particulate formation remain

Future Work

- PM characterization and filtration experiments
  - Extend EFA experiments to multiple filter substrates from many manufacturers spanning a wide range of properties
  - Third cooperative experimental campaign at U of Wisconsin ERC
    - Elucidate role of liquid fuel droplets in particulate formation by comparing particulates produced using pre-mixed and pre-vaporized fuel versus direct injection
    - Characterize the population of particles that are able to penetrate through filters at a variety of engine operating conditions
  - Work toward ability to conduct filtration experiments at realistic exhaust temperatures
  - Consider additional filtration experiments with simple surrogate particles
- Analysis and modeling
  - Analyze CT data from seven additional samples to support modeling of EFA experimental matrix
    - Three manufacturers
    - Three ceramic types
    - Wide range of porosity and pore size
  - Explore the use of Maximal Inscribed Sphere analysis at higher resolutions to connect 3D microstructural data with data from mercury porosimetry
  - Explore the use of Eulierian Lattice Boltzmann filtration simulations to improve device scale unit collector models
  - Continue development and validation of University of Wisconsin Heterogeneous Multi-scale Filtration model

Summary

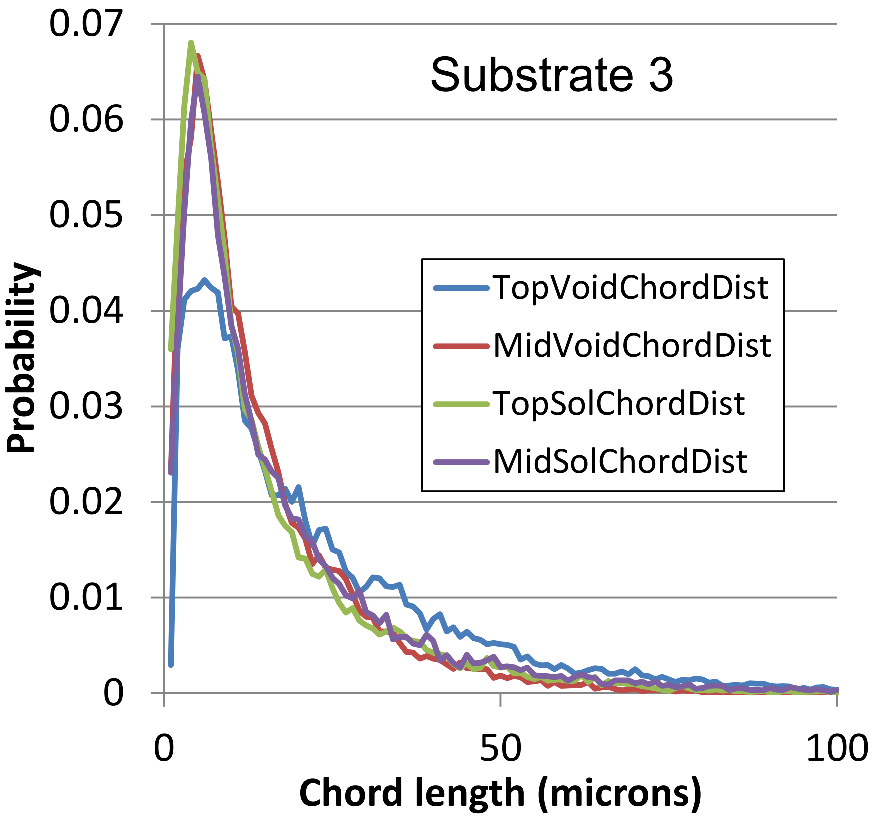
- Detailed characterization has been carried out for five current exhaust filtration substrates
  - Mercury porosimetry
  - Extensive high-resolution micro X-Ray CT data
    - Wall thickness
    - Porosity variation across walls
    - Autocorrelation
    - Chord length distribution
    - Maximal Inscribed Sphere analysis
    - Lattice-Boltzmann flow and filtration simulations
- The Heterogeneous Multi-scale Filtration (HMF) model was developed at the ERC for improved filter efficiency prediction across the particle size spectrum
- Initial filtration experiments were performed with the ERC Exhaust Filtration Analysis (EFA) platform and exhaust from a single-cylinder SIDI test engine

Technical Back-Up Slides

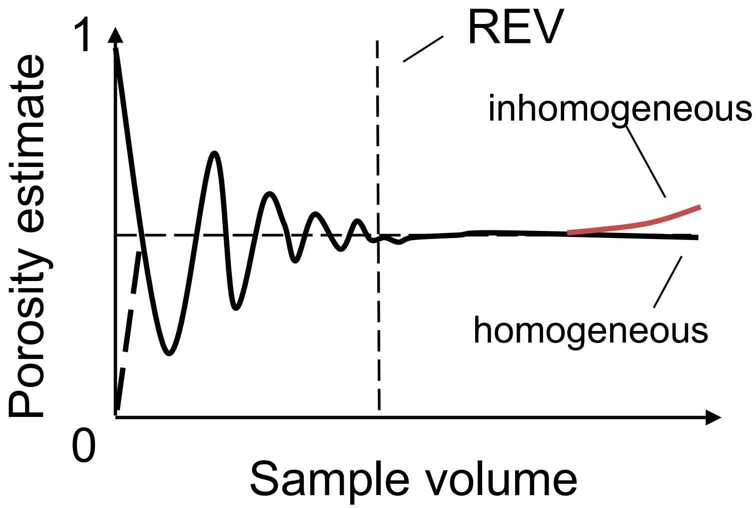
(Note: please include this “separator” slide if you are including back-up technical slides (maximum of five). These back-up technical slides will be available for your presentation and will be included in the DVD and Web PDF files released to the public.)

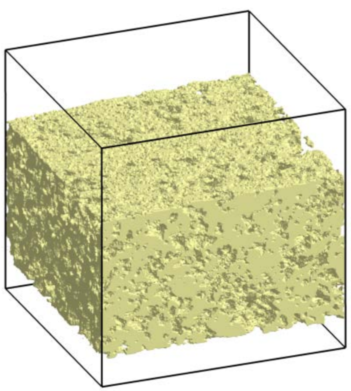
CT analysis - chord length distributions

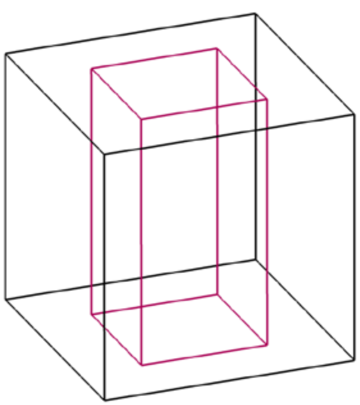
- Chord length distributions also encode information about length scales
- Peaks imply that we are capturing fine features of the structure
- They have been used as a "fingerprint" for digital reconstruction of porous media



Representative Elementary Volume (REV)







- Concept described by Jacob Bear in Dynamics of Fluids in Porous Media (1972)
- Accurate estimates of ‘macroscopic’ parameters can be made when the volume of material becomes large enough to average out variations in the porous medium
- Flow simulations were carried out with various sub-volumes of filter reconstructions with 3.2 micron resolution

Pore size distribution for HMF model

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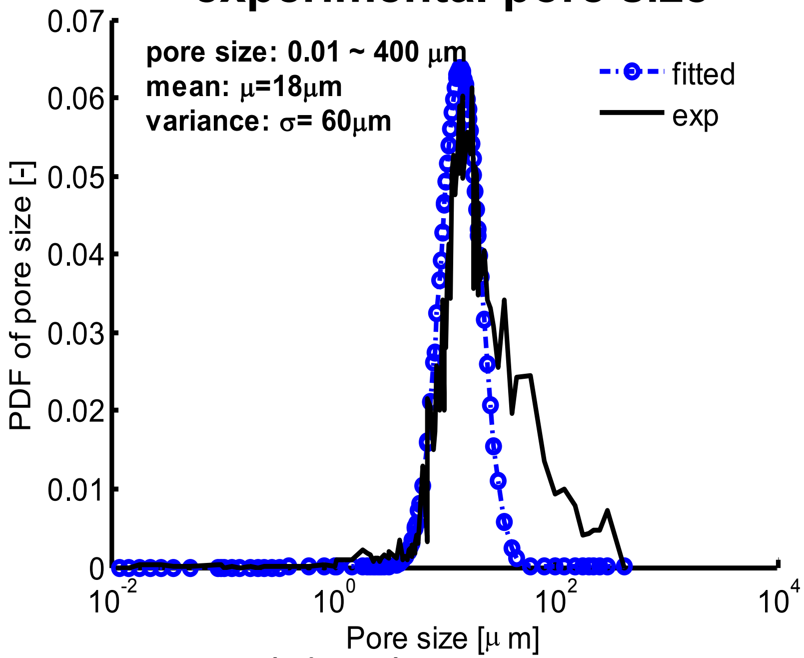
Fitted PDF using experimental pore size



- Lognormal distribution
  - Mean pore size
  - Variance

$$\int pdf_{dc_i} \cdot d(dc_i) = 1$$

$$\int pdf_{dc_i} \cdot dc_i \cdot d(dc_i) = dc_m$$

- The fitting criteria
  - nominal mean pore size “dp_max” is used
  - Give a variance
    - Represent the degree of how the pore size spreads out

→Symmetric pdf

- Utilize the most critical pore size
- Ignores some large size pre (relatively small pores contribute more on filtration efficiency)

EFA experimental layout

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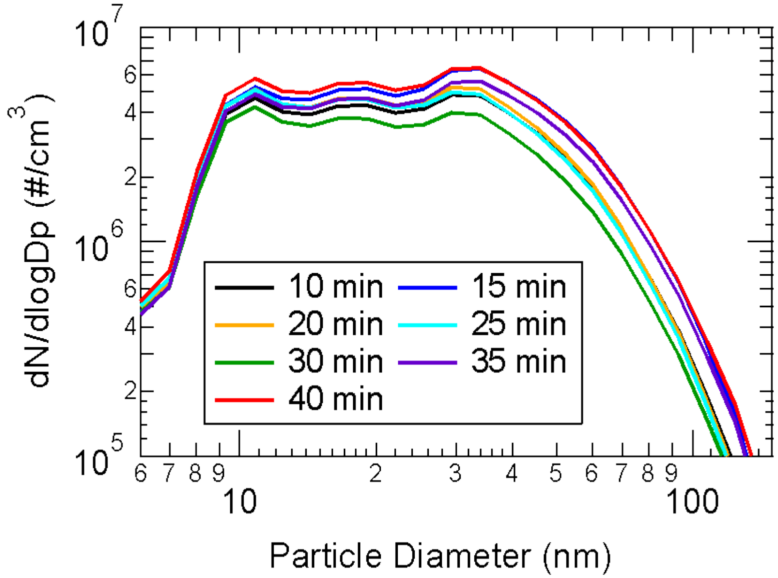
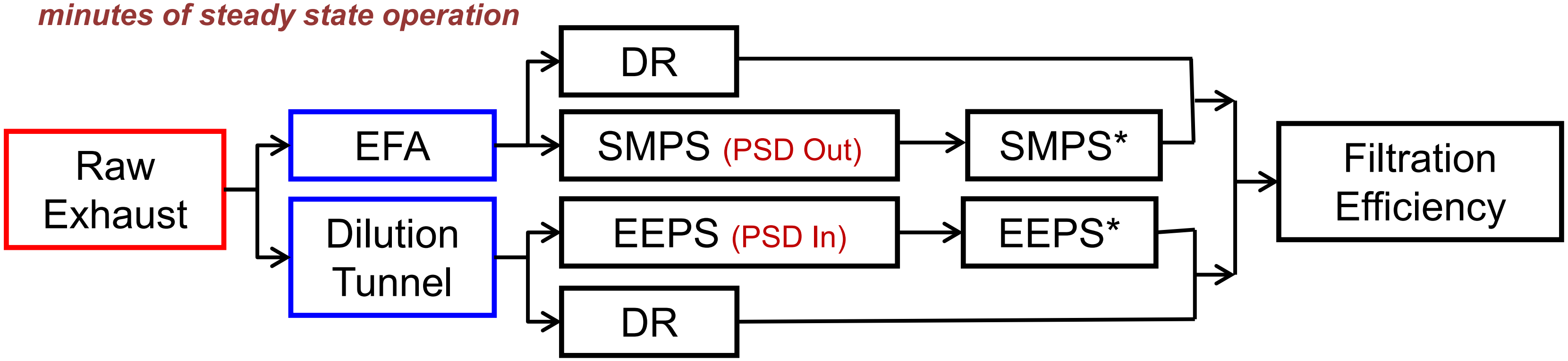


Change in PSD from the engine during 40 minutes of steady state operation

SMPS*
- Measurements converted to particle concentration versus time for each particle size
- Linear interpolation used reconstruct SMPS PSD at a 1 second resolution

EEPS*
- EEPS PSD corrected using the SMPS Ratio



See SAE-01-1158

EFA size resolved transient data

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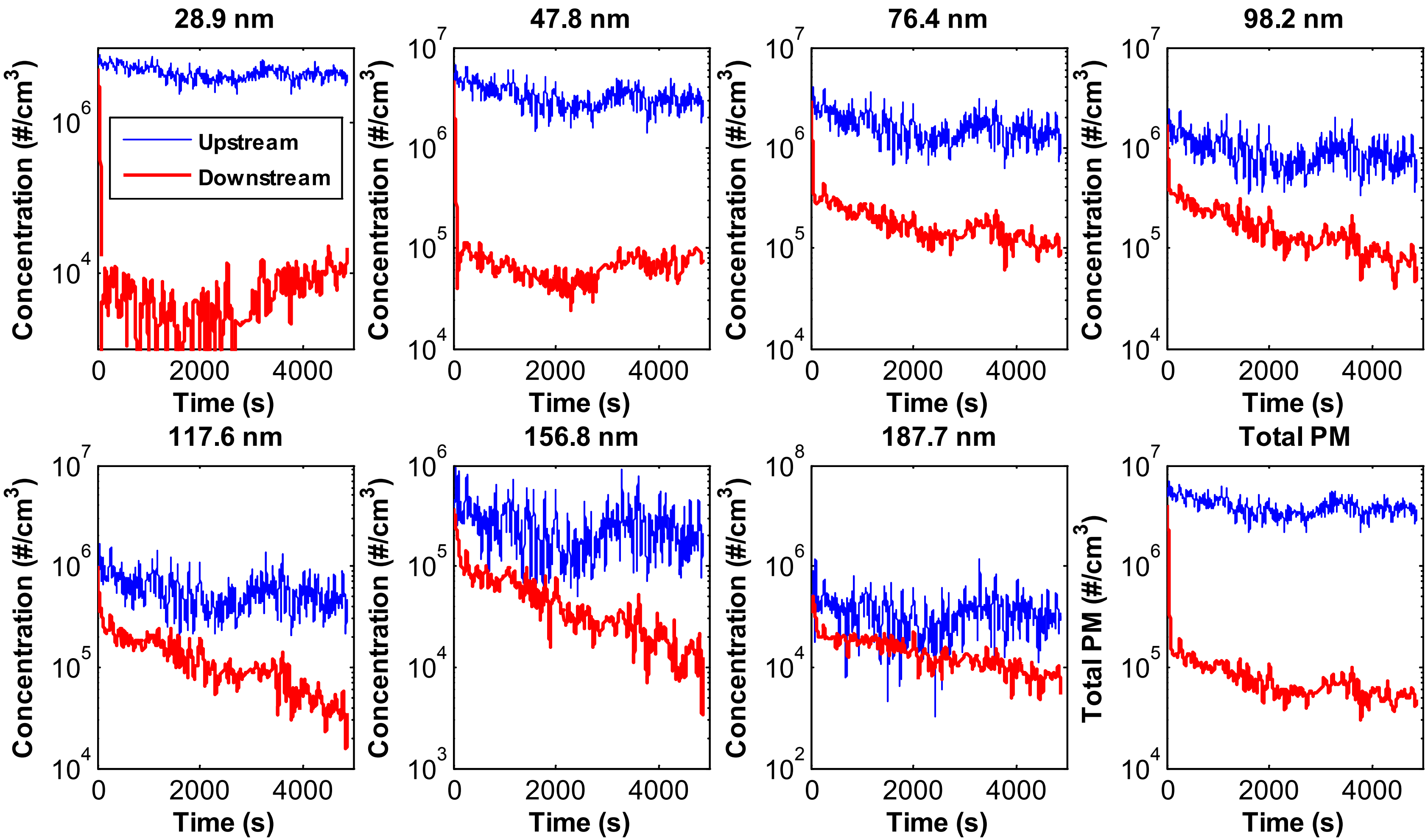

See SAE-01-1158